

Crossover from β to α relaxation in cooperative facilitation dynamics

Mauro Sellitto

*Dipartimento di Ingegneria Industriale e dell'Informazione, Seconda Università di Napoli,
Real Casa dell'Annunziata, Via Roma 29, I-81031 Aversa (CE), Italy*

β and α relaxation processes are dynamical scaling regimes of glassy systems occurring on two separate time scales which both diverge as the glass state is approached. We study here the crossover scaling from β - to α - relaxation in the cooperative facilitation scenario (CFS) and show that it is quantitatively described, with no adjustable parameter, by the leading order asymptotic formulas for scaling predicted by the mode-coupling theory (MCT). These results establish (i) the mutual universality of the MCT and CFS, and (ii) the existence of a purely dynamic realization of MCT which is distinct from the well-established random first order transition scenario for disordered systems. Some implications of the emerging kinetic-static duality are discussed.

The glass state is obtained with striking simplicity in a variety of substances [1]. Yet, its fundamental nature is one of the most enduring puzzles of condensed matter physics. From a dynamical point of view, what makes the glass relaxation so peculiar is the existence of *two* time scales which both grow dramatically as the separation σ from the glass state vanishes. The first scale, t_σ , refers to the rattling motion of particles in the cage formed by their neighbors (β relaxation). The second one, $t'_\sigma \gg t_\sigma$, is generally ascribed to the slower cooperative rearrangements of cages that allow long-term particle diffusion (α relaxation). This two-step relaxation pattern was first predicted by mode-coupling theory (MCT) [2], and entails as precursors phenomena strongly temperature or density dependent spectra exhibiting relaxation stretching. Pioneer work first identified these glass precursors in colloidal suspensions of hard spheres [3] and in molecular-dynamics simulations for a Lennard-Jones mixture [4].

The crossover from β to α relaxation, which occurs for times large on scale t_σ but small on scale t'_σ , constitutes a crucial test for our understanding of glassy dynamics [5]. It is remarkable because the ratio t'_σ/t_σ is itself diverging as $\sigma \rightarrow 0$. At the same time, however, its actual investigation is delicate because of the sensitive dependence on three quantities: the separation parameter σ , the system-specific exponent parameter λ , and the arrested part of correlation, f . In realistic systems these quantities can be inferred only with limited accuracy (or may even be not well defined). This makes it difficult to assess unambiguously the MCT predictions and only a few cases have been analyzed [6, 7]. In this Letter we provide a test of the crossover scaling in the cooperative facilitation scenario (CFS) [8]. Several results have already suggested a close analogy between MCT and CFS [9–12]. Here we show that this relation is, in fact, quantitative and deep.

MCT scaling laws. – Without loss of generality we refer to the ideal version of MCT, in which the normalized density-fluctuation correlator $\Phi(t)$ has no wave-vector dependence (for an exhaustive description of MCT, see Ref. [2]). Near the glass bifurcation singularity the correlator exhibits a plateau at some f_c . Near this plateau the small- σ dynamics on scale t_σ is ruled by the *first*

scaling law:

$$\Phi(t) - f_c = c_\sigma g(\hat{t}), \quad \hat{t} = \frac{t}{t_\sigma}. \quad (1)$$

Here the correlation scale c_σ and the time scale t_σ read

$$c_\sigma \propto \sqrt{|\sigma|}, \quad t_\sigma \propto |\sigma|^{-\frac{1}{2a}}, \quad (2)$$

where a is some critical exponent. The σ -independent master function $g(\hat{t})$ obeys the scale-invariant equation

$$-1 + \lambda g(\hat{t})^2 - \partial_{\hat{t}} \int_0^{\hat{t}} g(\hat{t} - \hat{t}') g(\hat{t}') d\hat{t}' = 0, \quad (3)$$

to be solved for the initial condition $\lim_{\hat{t} \rightarrow 0} g(\hat{t}) \hat{t}^a = 1$. The exponent parameter, λ , quantifies all properties of $g(\hat{t})$. In particular, it determines the exponent a via

$$\frac{\Gamma(1-a)^2}{\Gamma(1-2a)} = \lambda. \quad (4)$$

Two time regions can be distinguished in the first scaling regime where, depending on whether Φ is above or below f_c , the master function $g(\hat{t})$ can be approximated as

$$g(\hat{t}) \simeq \begin{cases} g_a(\hat{t}) = \hat{t}^{-a} - A_1 \hat{t}^a, & \text{for } \Phi > f_c \\ g_b(\hat{t}) = -B \hat{t}^b + \frac{B_1}{B} \hat{t}^{-b}, & \text{for } \Phi < f_c \end{cases} \quad (5)$$

with an error decreasing faster than \hat{t}^a in the former case and \hat{t}^{-b} in the latter. The coefficients A_1 and B_1 read

$$\frac{1}{2A_1} = \Gamma(1-a)\Gamma(1+a) - \lambda \quad (6)$$

$$\frac{1}{2B_1} = \Gamma(1-b)\Gamma(1+b) - \lambda, \quad (7)$$

while B is a positive quantity fixed only by λ . The dominant contribution to $g_b(\hat{t})$, $-B\hat{t}^b$, is called von Schweidler decay. The von Schweidler exponent b is determined by

$$\frac{\Gamma(1+b)^2}{\Gamma(1+2b)} = \lambda, \quad (8)$$

with $0 < b \leq 1$. The function $g(\hat{t})$ exhibits a zero at some \hat{t}^* . The part for \hat{t} preceding \hat{t}^* deals with deviation

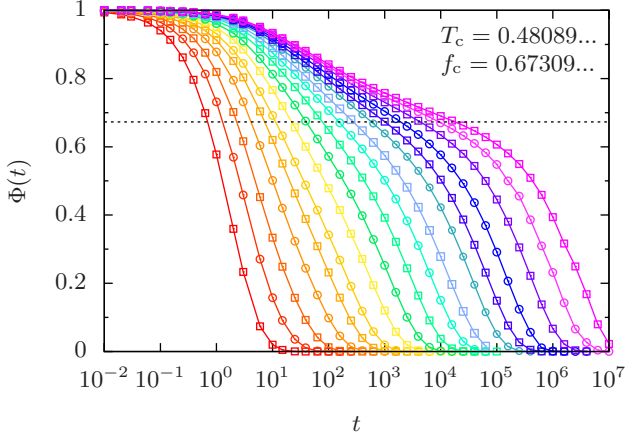


FIG. 1. Correlation decay for temperature $T \in [0.49, 10]$ in the CFS on a Bethe lattice with coordination $z = 4$ and facilitation parameter $f = 2$. System size $N = 2^{23-24}$.

from the critical decay. The part for $\hat{t} > \hat{t}^*$ describes the approach toward the von Schweidler decay. The quantities \hat{t}^* and B have to be calculated by solving Eq. (3). The numerical solution [5] shows that there is an interval for \hat{t} centered by \hat{t}^* , where the three functions g_a , g_b and g are very close to each other. Therefore, to locate approximately \hat{t}^* and B we set $g_a(\hat{t}^*) = g_b(\hat{t}^*) = 0$, which gives $\hat{t}^* \simeq A_1^{-1/2a}$ and $B \simeq \sqrt{A_1^{b/a} B_1}$. These approximations permit us to base the comparison with the CFS on elementary formulas.

Substituting the von Schweidler decay into Eq. (1), one gets the von Schweidler decay law

$$\Phi(t) - f_c = - \left(\frac{t}{t'_\sigma} \right)^b, \quad (9)$$

where the new relevant time scale is now

$$t'_\sigma \propto |\sigma|^{-\gamma}, \quad \gamma = \frac{1}{2a} + \frac{1}{2b}. \quad (10)$$

von Schweidler's law describes the small- σ dynamics for times intermediate between two diverging time scales, $t_\sigma \ll t \ll t'_\sigma$. It is important because of the connection it establishes between the late part of β relaxation to the early part of the α relaxation. The latter deals with the plateau-below dynamics on scale t'_σ and is globally governed by the *second scaling law* (also known as the time-temperature superposition principle):

$$\Phi(t) = \tilde{\Phi} \left(\frac{t}{t'_\sigma} \right). \quad (11)$$

Here $\tilde{\Phi}$ is another σ -independent master function. As an example, Figs. 1, 2 and 3 show the typical two-step relaxation and the associated MCT scaling laws for the CFS that we now describe.

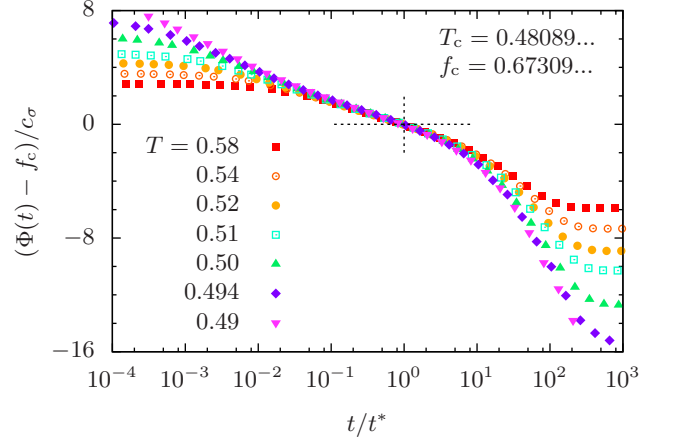


FIG. 2. MCT first scaling law in the CFS. Rescaled correlator vs rescaled time variable $\hat{t}/\hat{t}^* = t/t^*$.

CFS. – In the dynamic facilitation approach the mesoscopic structure of a liquid at temperature T is represented by an assembly of binary spins, $s_i = \pm 1$, $i = 1, \dots, N$, whose value depends on whether the local liquid density at site i is higher or lower than the average. Energetic interactions are absent but the spin dynamics is facilitated (or constrained) [8]: At each time step a randomly chosen spin s_i is flipped with probability $w(s_i \rightarrow -s_i) = \min\{1, e^{-s_i/T}\}$, provided that the spin s_i is surrounded by at least f nearby (liquidlike) up spins. This constraint mimics the cage effect: when the temperature is low enough the fraction of liquidlike spins is vanishingly small and, consequently, spin relaxation may involve a large number of cooperative spin flips over regions with increasing size. Here, the relevant range of the facilitation parameter, f , is $1 < f < z - 1$, where z is the lattice coordination. The correlator of our interest shall be the persistence $\Phi(t)$, i.e., the probability that a spin has never flipped between times 0 and t . Its long-time limit, the probability that a spin is permanently frozen, is precisely the analog of the nonergodicity parameter f . On the Bethe lattice this cooperative dynamics exhibits features qualitatively similar to MCT [9–13], though one would, naively, expect that the conventional form of MCT fails for this type of model as static correlations vanish. In fact, previous works on related facilitated systems on finite dimensional lattices showed that MCT-like approximations are unable to describe the overall glassy dynamics [14–16]. This can be generally understood through the connection of CFS with the bootstrap percolation transition which is smeared out in finite dimensions. For this reason, our evaluation of the MCT status is carried out on the Bethe lattice, which is the first natural step of a statistical mechanics treatment. This is also relevant because: (i) MCT scaling regimes are sometimes hardly observed in numerical simulations of disordered systems [20, 21], and (ii) MCT shows a be-

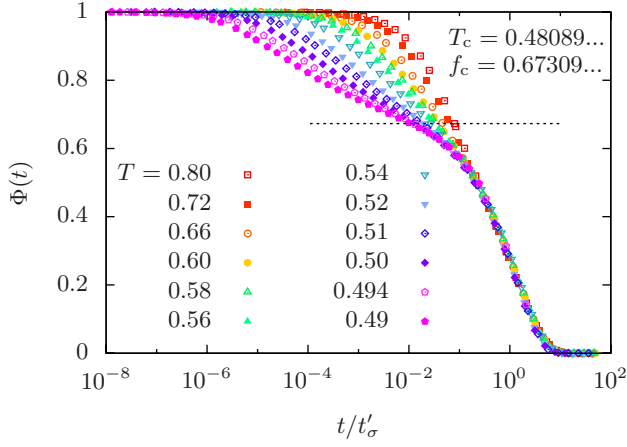


FIG. 3. MCT second scaling law in the CFS.

havior which, in the limit of large space dimensionality, does not conform to the replica theory [17–19].

To compare CFS with MCT we have first checked the β - and α -scaling laws along with Eqs. (2) and (10). We proceed as follows. The time scale t_σ is estimated as

$$t_\sigma = \int_0^{t^*} [\Phi(t) - \Phi(t^*)] dt, \quad (12)$$

where t^* is the time to cross the plateau, $\Phi(t^*) = f_c$. This is easily done as f_c is known exactly. For the Bethe lattice we consider here, $z = 4$ and $f = 2$, we have $T_c = 0.48089...$ and $f_c = 0.67309...$ [9]. Figure 4 shows that t_σ obeys the power law $t_\sigma \propto \sigma^{-1/2a}$ with an exponent $a \simeq 0.27$. This value is obtained by a fit over the temperature range [0.52, 0.82]. Notice, that a is the only critical exponent we estimate numerically here. The correlation scale c_σ , instead, is obtained from the MCT relation [2]:

$$c_\sigma = \sqrt{1 - \lambda} (f - f_c). \quad (13)$$

Here the jump of the order parameter, $f - f_c$, is deduced from the exact calculation on the Bethe lattice [9], while λ is estimated by exploiting Eq. (4) which gives $\lambda \simeq 0.815$. The result for the correlation scale c_σ is shown in Fig. 4 along with the MCT prediction. The excellent collapse of the rescaled relaxation data showed in Fig. 2 is obtained by using the c_σ calculated in this way. We then get $b \simeq 0.45$ and $\gamma \simeq 2.96$ from Eqs. (8) and (10). This latter value is consistent with that found in Ref. [9] ($\gamma \simeq 2.9$). As a consistency check we then estimate the time scale t'_σ through Eq. (12) with t^* such that $\Phi(t^*) = 0$. Figure 4 shows that t'_σ and the ratio t'_σ/t_σ are correctly predicted by MCT. From t'_σ we finally get the second scaling law for the α -relaxation process that we find to hold in a wide range of temperatures above T_c , see Fig. 3.

We now consider in detail the MCT predictions for the asymptotic leading order corrections to the critical decay laws, Eq. (5). The coefficients entering Eq. (5) as

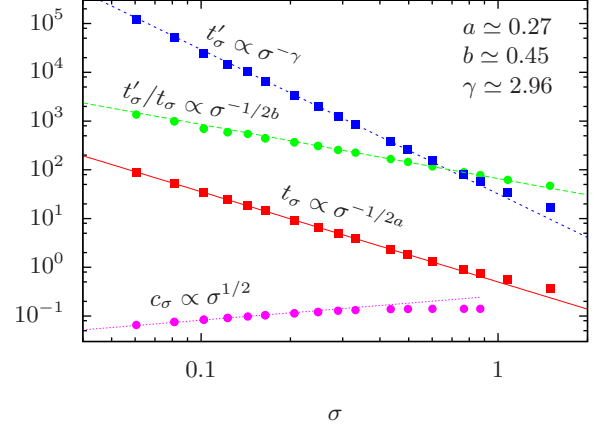


FIG. 4. Time scales t_σ and t'_σ and their ratio vs $\sigma = T/T_c - 1$ in the CFS. Data for the correlation scale c_σ are calculated exactly by using Eq. (13) with $\lambda \simeq 0.815$. Straight lines are MCT predictions.

obtained from the critical exponents estimated above are $A_1 \simeq 1.58$, $B_1 \simeq 0.812$ and $B \simeq 1.32$. We shall use these values to test in a self-consistent manner the MCT predictions: any observed deviation would mean that MCT does not hold for CFS either because the λ estimated for our system is not connected to the critical decay exponents a and b by Eqs. (4) and (8), or because Eqs. (5), (6) and (7) just do not apply to CFS. We have explored a relatively wide range of temperatures above T_c . Figure 5 shows the behavior of $\Phi(t)$ for some values of T along with the MCT predictions for the crossover through the plateau. The full line above f_c represents the function $f_c + c_\sigma g_a(\hat{t}/\hat{t}^*)$ while the plateau-below function corresponds to $f_c + c_\sigma g_b(\hat{t}/\hat{t}^*)$. To appreciate the effect of leading order corrections we also add the critical decay laws (which are obtained by setting $A_1 = B_1 = 0$) as dashed lines. We see that, in agreement with the nature of MCT corrections, the quality of comparison increases as T decreases. In particular, up to the 80% of the correlator shape is accurately reproduced, over a time window ranging from two to more than three decades; see Fig. 5. No significant improvement is achieved with the exact numerical solution of Eq. (3) [22]. It is interesting to observe that, even though MCT was not obviously devised for describing CFS, the MCT predictions perform even better than a recent truncation scheme for facilitated master equations; see Fig. 4 in Ref. [23] for a comparison. Thus, it seems that MCT fully captures in a very general way the slow dynamics of large scale cooperative rearrangements occurring near the glass singularity.

What makes the observed agreement pretty remarkable, however, is the absence of any fitting parameter in our procedure. In fact, one should keep in mind that the prefactors of leading order corrections, A_1 and B_1/B , are correlated and have no temperature dependence, i.e., they are strongly constrained. Thus, to check the sta-

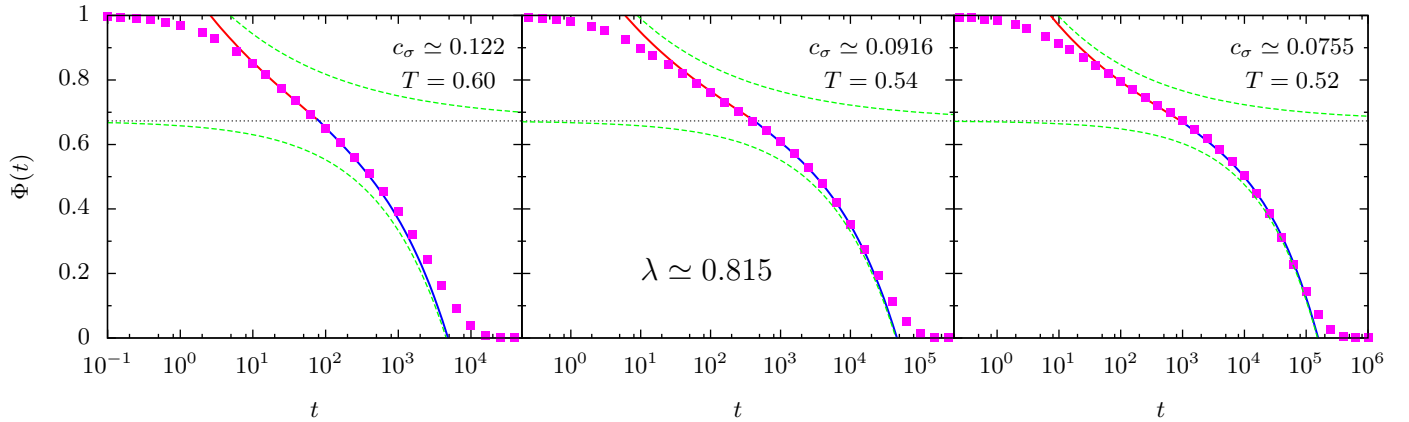


FIG. 5. Persistence $\Phi(t)$ vs time t at temperature T in the CFS. Full lines are the asymptotic leading corrections to the critical scaling laws predicted by MCT. The line above the plateau at $f_c = 0.67309$ represents the function $f_c + c_\sigma g_a(\hat{t}/\hat{t}^*)$ while the blue line below the plateau corresponds to $f_c + c_\sigma g_b(\hat{t}/\hat{t}^*)$. For comparison, pure critical decay laws are shown as dashed lines.

bility of our comparison, we changed the exponent a by an amount of ± 0.01 , that is essentially the uncertainty we have on this exponent. We then found that relaxation behavior is still well reproduced if all other quantities entering Eq. (5) are changed according to MCT relations. This robustness seems to suggest that prefactors of leading order corrections are rather optimal from the purely data fitting point of view. Nevertheless, we notice that very close to T_c (below $T = 0.51$), the theoretical value of c_σ need to be increased by a small factor (up to 5%) in order to accurately describe the intermediate stage of α relaxation. This discrepancy is presumably due to critical finite-size corrections which are hardly accounted for in a MCT description. The exact solution of a related cooperative facilitated system should help to settle this issue [24]. Since static correlations are absent here, we expect that a suitable MCT formulation of facilitation dynamics is related to those studied in Refs. [25, 26].

Conclusions. – We have shown that the relaxation behavior of cooperative facilitated spin models is accurately described, in a relatively wide range of temperatures, by the MCT predictions for the asymptotic leading corrections to critical decay laws. These results support the idea of glassy universality and put on a solid quantitative ground the correspondence between MCT and CFS. At the same time new challenges arise.

It is well known that the dynamics of disordered p-spin-like systems is also exactly described by MCT which is the key to the celebrated random-first order transition (RFOT) scenario for the glass transition [27, 28]. RFOT is based on a nontrivial phase structure characterized by a one-step replica symmetry breaking and a possible fractal free-energy landscape lying deeper in the glass phase [29]. In the CFS, instead, thermodynamics plays no role: the configuration space breaks into several components that cannot be connected by a sequence of allowed

spin flips. The duality between kinetic and static representations seems therefore irreconcilable. This would imply, for example, that the dynamical inverse problem of inferring the interactions among system units starting solely from time correlation data is undecidable. However, in view of the double connection (between MCT and CFS on one hand, and between MCT and RFOT on the other) one is led to conclude that a map connecting CFS and RFOT must necessarily exist [30]. Extending the mapping below the Kauzmann temperature would obviously require extra thermodynamic ingredients which are arguably provided by suitable energetic interactions, as shown in Ref. [31]. Thus, the existence of three *a priori* distinct frameworks that give the same dynamical scaling laws, while certainly surprising, should not be considered as a mere coincidence, but rather as a genuine signature of the universality of glassy relaxation captured by these frameworks. One advantage of the CFS is that the glass formation can be interpreted in real space as the formation of a bootstrap percolation backbone of permanently frozen spins. Much less clear is how to extract the values of the critical exponents a and b from this geometrical structure, since no spin is permanently frozen on the liquid side of the glass transition. So, at the moment this important issue remains elusive in this framework as well as in other ones. Another perspective suggested by the CFS is that the ergodicity-restoring activated events in finite dimensions should be interpreted as the analog of the finite-volume metastability effects which are known to transform the bootstrap percolation transition in a (sharp) crossover [32]. Finally, it would be crucial to extend the above investigation to finite dimensional facilitated systems having a glass transition (see, e.g., Ref. [33]). The exploration of these problems is left to future works.

W. Götze is gratefully acknowledged for elucidations about MCT and for his interest in this work. I also thank S. Franz and R. Schilling for valuable comments on the manuscript.

-
- [1] K. Binder and W. Kob, *Glassy Materials and Disordered Solids* (World Scientific, Singapore 2011).
 - [2] W. Götze, *Complex Dynamics of Glass-Forming Liquids* (Oxford University Press, Oxford, 2009).
 - [3] W. van Meegen, S.M. Underwood and P.N. Pusey, Phys. Rev. Lett. **67**, 1586 (1991).
 - [4] W. Kob and H.C. Andersen, Phys. Rev. Lett. **73**, 1376 (1994).
 - [5] W. Götze, J. Phys. Condens. Matter **2**, 8485 (1990).
 - [6] T. Franosch, M. Fuchs, W. Götze, M.R. Mayr, and A.P. Singh, Phys. Rev. E **55**, 7153 (1997).
 - [7] T. Gleim and W. Kob, Eur. Phys. J. B **13**, 83 (2000).
 - [8] G.H. Fredrickson and H.C. Andersen, Phys. Rev. Lett. **53**, 1244 (1984); J. Chem. Phys. **83**, 5822 (1985).
 - [9] M. Sellitto, G. Biroli, and C. Toninelli, Europhys. Lett. **69**, 496 (2005).
 - [10] M. Sellitto, D. De Martino, F. Caccioli, and J.J. Arenzon, Phys. Rev. Lett. **105**, 265704 (2010); J.J. Arenzon and M. Sellitto, J. Chem. Phys. **137**, 084501 (2012).
 - [11] M. Sellitto, Phys. Rev. E **86**, 030502(R) (2012); J. Chem. Phys. **138**, 224507 (2013).
 - [12] S. Franz and M. Sellitto, J. Stat. Mech. (2013) P02025.
 - [13] H. Ikeda and K. Miyazaki, Europhys. Lett. **112** 16001 (2015).
 - [14] K. Kawasaki, Physica A (Amsterdam) **215**, 61 (1995).
 - [15] S.J. Pitts, T. Young, and H.C. Andersen, J. Chem. Phys. **113**, 8671 (2000).
 - [16] M. Einax and M. Schulz, J. Chem. Phys. **115**, 2282 (2001).
 - [17] A. Ikeda and K. Miyazaki, Phys. Rev. Lett. **104**, 255704 (2010); **106**, 049602 (2011).
 - [18] B. Schmid and R. Schilling, Phys. Rev. E **81**, 041502 (2010); R. Schilling and B. Schmid, Phys. Rev. Lett. **106**, 049601 (2011).
 - [19] P. Charbonneau, A. Ikeda, G. Parisi, and F. Zamponi, Phys. Rev. Lett. **107**, 185702 (2011).
 - [20] C. Brangian, W. Kob, and K. Binder, J. Phys. A **35**, 191 (2002).
 - [21] T. Sarlat, A. Billoire, G. Biroli, and J.-P. Bouchaud, J. Stat. Mech. P08014 (2009).
 - [22] The numerical code for solving Eq. (3) has been kindly provided by Th. Voigtmann.
 - [23] P.G. Fennell, J.P. Gleeson, and D. Cellai, Phys. Rev. E **90**, 032824 (2014).
 - [24] G. Parisi and M. Sellitto, Europhys. Lett. **109**, 36001 (2015).
 - [25] K. Kawasaki and B. Kim, Phys. Rev. Lett. **86**, 3582 (2001); J. Phys. Condens. Matter **14**, 2265 (2002).
 - [26] R. Schilling and G. Szamel, Europhys. Lett. **61**, 207 (2003); J. Phys. Condens. Matter **15**, S967 (2003).
 - [27] V. Lubchenko and P.G. Wolynes, Annu. Rev. Phys. Chem. **58**, 235 (2007).
 - [28] T.R. Kirkpatrick and D. Thirumalai, Rev. Mod. Phys. **87**, 183 (2015).
 - [29] P. Charbonneau, J. Kurchan, G. Parisi, P. Urbani, and F. Zamponi, Nat. Commun. **5**, 3725 (2014).
 - [30] For a relatively simpler case in which this mapping can be effectively realized, see L. Foini, F. Krzakala, and F. Zamponi, J. Stat. Mech. (2012) P06013.
 - [31] S. Franz, G. Gradenigo, and S. Spigler, arXiv:1507.05072
 - [32] M. Aizenman and J. L. Lebowitz, J. Phys. A **21**, 3801 (1988).
 - [33] H. Ohta and S. Sasa, J. Stat. Phys. **155**, 827 (2014).